Schorl composition from the barren granitic pegmatites in the Western Carpathians: Two examples from the Nízké Tatry Mountains, Central Slovakia

Složení skorylu z primitivních granitických pegmatitů Západních Karpat: dva příklady z Nízkých Tater, střední Slovensko (Czech summary)

(2 figs)

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Tourmaline group minerals belong to rather uncommon accessory minerals of the Hercynian granitic pegmatites of the Central Western Carpathians, Slovakia. Black tourmaline from two barren quartz-feldspar-(biotite-muscovite) pegmatites of the Nízké Tatry Mts.: Bystrá Valley – Srdiečko (TBV) and Magurka – Adolf adit (TMA), were studied by EMPA. Both tourmalines show schorl compositions with intermediate X-site vacanies: 0.25–0.33 *pfu*, slight compositional zonality with increasing Fe/(Fe+Mg) ratio from center to rim of crystals, and low Ca and K contents. The most striking feature of the TMA schorl is a notably high Mn content: 0.98–1.56 wt. % MnO, 0.14–0.23 *apfu*, exceptional for schorl from poorly fractionated granite-pegmatite environment. Nevertheless, the TMA schorl also reveals higher Fe/(Fe+Mg) ratio in comparison to TBV: 0.69–0.83 and 0.54–0.60, respectively, and Mn values correlate positively with Fe, Fe/(Fe+Mg) and X-site vacancies. Thus, the elevated Mn content in TMA schorl is probably a consequence of local Mn-rich environment, probably due to absence of garnet as well as a relatively higher overall fractionation level.

Key words: schorl, tourmaline, barren granitic pegmatite, Nízké Tatry Mts., Slovakia

Introduction

Minerals of the tourmaline group do not belong to the typical accessory phases of Hercynian, mainly Carboniferous granitic rocks and their pegmatites of the Tatric and Veporic Superunits in the West-Carpathian orogenic belt, in contrast to the abundance of tourmalines in evolved tin-bearing Permian leucogranites of the Gemeric Superunit (e. g. Faryad – Jakabská 1996, Broska et al. 1998, 1999).

Scarce occurrences of accessory tourmaline in Tatric and Veporic pegmatites have been reported from the Kondracká Kopa Saddle and Goričková Hill, Western Tatry Mts. (Pawlica 1914 in Koděra et al. 1990), near Sklené village, Žiar Mts. (Fiala 1931), in several occurrences of the Bratislava area, Malé Karpaty Mts. (Valach 1954, Veselský – Gbelský 1978), at the Dubná Skala quarry near Martin, Malá Fatra Mts. (Ivanov – Kamenický 1957, Láznička 1965), in the Bystrá Valley, Nízké Tatry Mts. (Turan 1961), near Rejdová village, Slovenské Rudohorie Mts. (Chovan – Határ 1978) and in the vicinity of Čierny Balog and České Brezovo villages, Slovenské Rudohorie Mts. (Ženiš – Hvožďara 1985). All of the above-mentioned tourmaline localities belong to small primitive and barren granitic pegmatites without rare-element mineralization. Thus, the mineralogy and petrology of these "uninteresting" pegmatites, especially the composition of minerals, is poorly known to date; only two chemical compositions of West-Carpathian pegmatite tourmalines have been published many years ago (Pawlica 1914 in Koděra 1990) and they are not reliable. Moreover, the vast majority of recent papers dealing with pegmatite tourmalines described compositions especially from highly-evolved complex Li-Ta-Cs-rich granitic pegmatites. In an effort to partly fill this gap, we contribute new data on tourmaline compositions from two barren granitic pegmatites in the Nízké Tatry Mts., Central Slovakia.

Geology

Both localities of tourmaline described here are situated in the Tatric crystalline complex of the Nízké Tatry Mts., Central Western Carpathians. The area consists of Proterozoic (?) to Lower Paleozoic metapelitic-metapsammitic and metaigneous rocks, mainly biotite and two-mica sillimanite(±garnet±cordierite)-bearing paragneisses and orthogneisses, locally migmatitised (Krist et al. 1992, Biely – Bezák 1997). Orthogneisses with porphyritic K-feldspar are recently interpreted as Early Paleozoic granitic rocks overprinted during Hercynian orogenesis (Petrík et al. 1998). Less frequent are lenses of graphite-bearing quartz paragneisses, amphibole gneisses and amphibolites. Meta-ultramafic rocks and biotite-diopside gneisses, biotite phyllites to phyllitic mica schists, metavolcaniclastic lithologies, siderite-ankerite-muscovite and quartz-muscovite metasandstones rarely occur at local scale (Koutek 1931, Krist et al. 1992, Korikovsky – Molák 1995, Biely – Bezák 1997, Molák et al. 1998). The P-T conditions of Hercynian metamorphism attained 610–690 °C, 400–470 MPa in paragneisses and orthogneisses, and 320–330 to 450–480 °C, ≥350 MPa in metasandstones to phyllites and mica schists (Korikovsky – Molák 1995, Krist et al. 1992, Biely – Bezák 1997).

The metamorphic rocks are intruded by Hercynian granitic rocks: the principal varieties are the Ďumbier biotite tonalites to granodiorites and comagmatic Prašivá muscovite-biotite granodiorites to granites, with pinkish K-feldspar phenocrysts; less frequent are amphibole-biotite diorites and leucocratic granites (Koutek 1931, Lukáčik 1981, Biely – Bezák 1997). The Ďumbier-Prašivá pluton displays calc-alkaline transitional I-S-type trend (Kohút 1998) typical for the majority of the Hercynian synorogenic crustal intrusions in Tatric and Veporic Superunits of the Western Carpathians. Rb-Sr isotope dating of the pluton yielded a wall-rock isochron age of 368±22 Ma and IR_{Sr} = 0.7078(4) (Bagdasaryan et al. 1985) which documents the Lower Carboniferous age of the intrusion and the crustal character of the protolith.

Pegmatites

The Bystrá Valley, Srdiečko pegmatite (TBV) occurs in an outcrop at a forest road about 250 m NW from the Srdiečko Hotel and about 9 km N from Bystrá village, at an elevation of 1,180 m a. s. l. on the southern slope of the Nízké Tatry Mts. The Ďumbier biotite tonalite to granodiorite is the host rock of the pegmatite; the contact between the pegmatite and granite is sharp. Biotite to two-mica orthogneisses with banded structure are situated in close vicinity. The thickness of the pegmatite vein is around 1 m. The TBV pegmatite is slightly zoned with a gradual coarsening of minerals from wall to core. The mineral composition is simple: pinkish K-feldspar, white quartz and less frequent biotite and muscovite. The size of rock-forming minerals varies between 0.5 to 5 cm. Black tourmaline forms crystals 0.5 to 4 cm in lenght, enclosed in coarse-crystalline quartz-K-feldspar matrix in the central part of the pegmatite body.

Tourmaline-bearing granitic pegmatites in the Bystrá Valley area were mentioned by Turan (1961) and Láznička (1965), and K-feldspar from the TBV pegmatite was determined as maximum microcline (Dávidová 1978).

The Magurka, Adolf adit pegmatite (TMA) was found only as a rock fragment on dumps of the Adolf adit in the area of the Magurka gold-antimony ore deposit, ca. 600 m S from the Magurka settlement, at an elevation of 1,170 m a. s. l. on N slope of the Nízké Tatry Mts. The Prašivá biotite granodiorite to granite is the host rock of the pegmatites and hydrothermal quartz-sulphide veins. The fragment of pegmatite (ca. 7 cm in size) contains subhedral aggregate of white-grey quartz, grey partly sericitized plagioclase (albite to oligoclase) and slightly pinkish K-feldspar; the size of minerals varies between 3 and 7 mm. Black tourmaline forms a cluster of several small crystals, up to 5 mm in size. Scarce pyrite and stibnite (0.5–1 mm in size) rim and partly penetrate the tourmaline. They represent a younger hydrothermal overprint of the pegmatites.

Experimental methods

Electron microprobe analysis (EMPA) of tourmaline was done in energy-dispersion (EDS) mode on JEOL JCXA 733-Superprobe instrument with KEVEX system at Geological Survey of Slovak Republic, Bratislava. The beam diameter was 3–5 μm, accelerating potential 15 kV, beam current around 1200 nA and sample counting time 100 s. Synthetic materials and natural minerals were used as standards for Kα X-ray lines. ZAF normalisation was used. The lower detection limit for all elements is 0.05–0.10 wt. %, the error in EMPA determination is in the range of ±0.1 to 0.3 wt. %. The structural formulae were calculated on the basis of sum T+Z+Y cations = 15 assuming OH^- = 4, and B = 3 atoms per formula units (*apfu*).

Results

Composition of the Bystrá Valley, Srdiečko tourmaline (TBV)

Tourmaline is homogeneous in BSE images. One profile through (0001) plane of an about 6 mm crystal section and across two additional smaller oblique crystal sections, was investigated (9 analyses in total). EMPA study shows a relatively homogeneous schorl composition of the tourmaline (Table 1, anal. 1–4, Fig. 1). TBV schorl is relatively rich in Al: total Al = 6.65–6.82 *apfu*, ^{T}Al (T-site Al) = 0.07–0.21 *apfu* and ^{Y}Al = 0.44–0.74 *apfu*, but low in Ti (0–0.045 *apfu*, 0–0.4 wt. % TiO_2) and especially Mn (below detection limit). Fe/(Fe+Mg) atomic ratio is between 0.54 to 0.60; thus, the tourmaline represents a Mg-rich schorl. Na strongly prevails among the X-site elements, Ca and K contents are negligible: 0.18–0.39 wt. % CaO (0.03–0.07 Ca *apfu*) and 0.00–0.08 wt. % K_2O (near detection limit, up to 0.02 K *apfu*). X-site vacancies show intermediate values, 0.22–0.37 *pfu*. The center-to-rim trends in crystals are only slightly developed: Fe increases (1.25 to 1.49 *apfu*), Mg is stable or decreases (1.10 to 0.96 *apfu*) and ^{Y}Al commonly decreases (0.74 to 0.44 *apfu*). The differences in all other elements are negligible.

Composition of the Magurka, Adolf adit tourmaline (TMA)

Tourmaline from TMA also shows homogeneous BSE images. Three crystals were investigated by EMPA in profiles from center to rim of an oblique section nearly perpendicular to the (0001) plane; a total of 10 point analyses were done. Microprobe analysis reveals moderately heterogeneous schorl compositions (Table 1, Fig. 1). The Al contents are slightly lower than in TBV schorl but still relatively elevated: total Al = 6.28–

Table 1 Representative compositions of TBV and TMA schorl (oxides in wt. %). 1, 2: number of crystal; C: center, I: intermediate, R: rim of crystals.
* calculated on the basic of ideal stoichiometry.

Anal. #	TBV-1-C	TBV-1-I	TBV-1-I	TBV-1-R	TMA-1-C	TMA-1-I	TMA-1-R	TMA-2-R
SiO_2	35.53	35.37	35.41	35.31	34.34	34.43	34.60	34.26
TiO_2	0.00	0.18	0.00	0.28	0.33	0.34	0.33	0.10
B_2O_3 *	10.41	10.42	10.42	10.58	10.19	10.33	10.37	10.27
Al_2O_3	34.59	34.68	34.42	34.37	31.24	32.06	32.65	31.65
FeO_{tot}	8.99	9.18	9.68	10.77	12.87	15.11	14.59	15.46
MnO	0.00	0.11	0.00	0.00	0.98	1.48	1.45	1.58
MgO	4.03	3.89	3.93	4.20	3.32	1.85	1.86	1.81
CaO	0.18	0.18	0.20	0.32	0.14	0.09	0.15	0.23
Na_2O	1.98	2.09	1.89	1.98	2.13	2.14	2.01	1.87
K_2O	0.06	0.07	0.08	0.00	0.07	0.00	0.06	0.07
H_2O *	3.59	3.60	3.60	3.65	3.52	3.56	3.58	3.54
Total	99.36	99.77	99.63	101.46	99.13	101.39	101.65	100.84
Atomic proportions based on the sum of T+Z+Y = 15 cations								
Si	5.933	5.898	5.906	5.801	5.857	5.795	5.797	5.800
^{T}Al	0.067	0.102	0.094	0.199	0.143	0.205	0.203	0.200
Total *T*	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000
B	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
Al *Z*	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000
Ti	0.000	0.023	0.000	0.035	0.042	0.043	0.042	0.013
^{Y}Al	0.741	0.714	0.672	0.456	0.137	0.155	0.244	0.115
$Fe^{2+,3+}$	1.256	1.280	1.351	1.480	1.835	2.127	2.043	2.188
Mn	0.000	0.016	0.000	0.000	0.142	0.211	0.206	0.227
Mg	1.003	0.967	0.977	1.029	0.844	0.464	0.465	0.457
Total *Y*	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
Total Al	6.808	6.816	6.766	6.655	6.280	6.360	6.447	6.315
Ca	0.032	0.032	0.036	0.056	0.026	0.016	0.027	0.042
Na	0.641	0.676	0.611	0.631	0.704	0.698	0.653	0.614
K	0.013	0.015	0.017	0.000	0.015	0.000	0.013	0.015
Total *X*	0.686	0.723	0.664	0.687	0.745	0.714	0.693	0.671
Vac. *X*	0.314	0.277	0.336	0.313	0.255	0.286	0.307	0.329
Total Cat.	18.686	18.723	18.664	18.687	18.746	18.714	18.694	18.672
OH	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000
O	31.196	31.207	31.139	31.035	30.924	30.883	30.922	30.826
Mn/(Mn+Fe)	0.000	0.012	0.000	0.000	0.072	0.090	0.091	0.094
Fe/(Fe+Mg)	0.556	0.570	0.580	0.590	0.685	0.821	0.815	0.827

6.46 *apfu*, ^{T}Al = 0.14–0.26 *apfu* and ^{Y}Al = 0.14–0.24 *apfu*. Ti contents are also very similar to TBV: 0.1–0.4 wt. % TiO_2 (0.01–0.05 Ti *apfu*). However, Fe/(Fe+Mg) of 0.69–0.83 is significantly higher than in the TBV schorl. The X-site elements display a pattern similar to that of TBV: dominant Na, low Ca (0.08–0.23 wt. % CaO, 0.01–0.04 Ca *apfu*) and K (around the detection limit, up to 0.07 wt. % K_2O and up to 0.015 K *apfu*), as well as intermediate level of X-site vacancy (0.25–0.33). However, the most notable geochemical feature of the TMA schorl is the strikingly high Mn content, 0.98 to 1.56 wt. % MnO, 0.14–0.23 Mn *apfu*. Manganese increases from the center to the rim of crystals, from 0.14 to 0.23 Mn *apfu*; it positively correlates with Fe, Fe/(Fe+Mg) and X-site vacancy value, and negatively correlates with Mg (Fig. 2A–D). Fe also increases from center to rim (1.84 to 2.19 Fe *apfu*), in contrast to decreasing Mg (from 0.85 to 0.47 Mg *apfu*), which results in an increase in the Fe/ /(Fe+Mg) ratio from 0.69 in center to 0.83 in rim of crystals. Despite the positive correlation between Mn and Fe, Mn/(Mn+Fe) (at.) also increases from the center (0.072) to rim (0.094), and Fe/Mn decreases from 12.97 to 9.67. Finally, correlations between Mg and Fe, as well as Mg and Fe+Mn are negative (Fig. 2E, F).

Consequently, at least simple $MnMg_{-1}$, and $FeMg_{-1}$ and $MnFe_{-1}$ substitutions could be expected. The Si and Al contents of the TMA schorl are relatively stable, and Na slightly decreases toward the rim (from 0.70 to 0.61 Na *apfu*).

Discussion

Tourmaline-group minerals are very sensitive petrologic indicators due to their complex structure and a broad scale of cation and anion substitutions (Hawthorne – Henry 1999). Granitic pegmatites are typical examples of compositional variability and evolutional trends of tourmalines from primitive barren to the most complex rare-element pegmatites. The level of fractionation, P-T parameters, oxygen and fluorine fugacity as well as local geochemical environments (e. g. Ca-rich host rocks) have an important influence on chemical composition and zonality of pegmatite tourmalines (e. g. Novák 1998, Bilal et al. 1998, Novák et al. 1999, Selway et al. 1999).

Both the TBV and TMA tourmalines have schorl composition and indicate a primitive barren nature of the parental granitic pegmatites; however, they also exhibit some differences. Generally, they are comparable with common Al-, Na-rich and Ca-poor schorl to dravite compositions from barren granitic pegmatites (Povondra 1981). The TBV schorl clearly shows a low level of magmatic fractionation and relatively simple evolution of host pegmatite: it is a Mg-rich schorl only slightly enriched in Fe along the crystals rims. Very low contents up to virtual absence of Mn also support this evaluation. On the contrary, the TMA schorl reveals a relatively higher fractionation level and more distinctive evolutionary

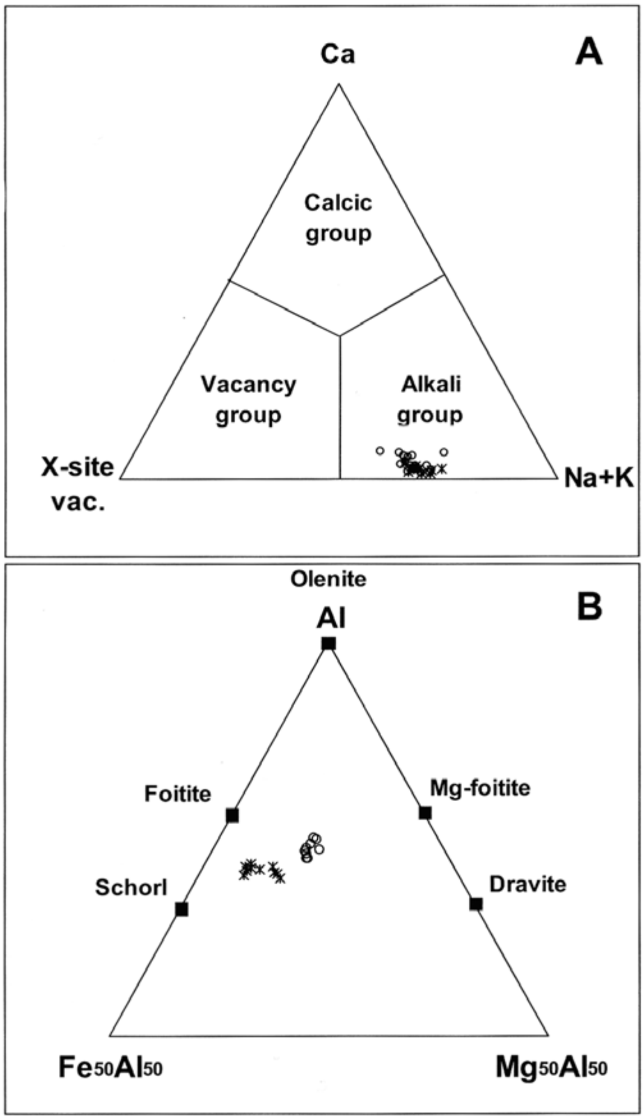

Fig. 1 Composition of the Bystrá Valley, Srdiečko (circles) and Magurka, Adolf adit (stars) schorl in ternary (A) Ca – X-site vacancy – Na+K and (B) Al-Fe-Mg diagrams (atomic proportions).

trend of the pegmatite: the mineral is Fe-rich with gradual Fe/(Fe+Mg) increase from center to rim of crystals. The high Mn content is the most strinking feature of the TMA schorl; it is higher than that in the analogous schorl from barren granitic and pegmatite environments, where Mn content is commonly less than 0.5 wt. %, only exceptionally up to 1 wt. % MnO (e. g. Shmakin 1976, Povondra 1981, Povondra et al. 1998, Broska et al. 1998, Pivec et al. 1998, Novák 1998). On the other hand, Li-rich schorl and especially elbaite contains ~1 to 8.6 wt. % MnO (Aurisicchio – Pezzotta 1997, Novák et al. 1999, Selway et al. 1999, Federico et al. 1998, Bilal et al. 1998). The high Mn content in tourmaline of these Li-rich complex rare-element pegmatites is caused by (i) extreme fractionation of the pegmatite, (ii) crystal-chemical reasons, (iii) specific geochemical environment, (iv) absence of other Mn-rich phase such as spessartine-rich garnet (Aurisicchio – Pezzotta 1997, Novák et al., in press) or a combination of (i) to (iv). All geological and mineralogical features preclude the existence of more-fractionated complex granitic pegmatites in the Magurka area. Moreover, despite the lack of ion-microprobe or another exact determination of Li in the TMA schorl, the stoichiometry of EMPA-based compositions indicates negligible, if any, Li content. Thus, the exceptionally high Mn content is probably caused by specific Mn-rich local geochemical environment; possibly, contamination by some Mn-rich mineral from the host rock could play a role. On the other hand, Fe/Mn of tourmaline decreases with parental pegmatite fractionation; this ratio has been used as a qualitative index of pegmatite fractionation (Bilal et al. 1998), similarly as in garnet and columbite-tantalite. Actually, the negative correlation between Mn and Mg, as well as a slight enrichment in Mn and increasing Mn/(Mn+Fe) also indicate the role of Mn fractionaction during crystallization of the TMA schorl. The Mn content of tourmaline is also strongly controlled by the association with spessartine-rich garnet (Aurisicchio – Pezzotta 1997, Novák et al., in press); consequently, the absence of garnet in the TMA pegmatite sample could also be a supporting factor for crystallization of Mn-bearing schorl. Unfortunately, our observation is limited to a single, tourmaline-bearing and garnet-free fragment, and to additional pegmatite fragments, which contain neither tourmaline nor garnet. The exposure of the pegmatite is not accessible. Consequently, only tentative conclusions can be formulated: the elevated Mn content in the TMA schorl is probably a consequence of local Mn-rich crystallization environment, possibly in absence of garnet as well as at a relatively high fractionation level of parental pegmatite.

Conclusions

Electron-microprobe study of tourmaline from two barren granitic pegmatites from the Nízké Tatry Mountains, Central Slovakia gave the following results:

(1) The studied tourmaline samples show schorl compositions, rich in Al (mainly TBV), with medium to relatively high Mg, Fe/(Fe+Mg) = 0.54–0.60 and 0.69–0.83, low Ti, Ca and K and medium X-site vacancies (~20–30 atom. %).

(2) Schorl crystals show slight compositional zonality with increasing Fe and Fe/(Fe+Mg) from center to rim.

(3) Exceptionaly high Mn content in the TMA schorl, uncommon for barren granite-pegmatite suites, as well as an increase in Mn/(Mn+Fe), Fe and decrease in Mg toward the crystal rim probably document the influence of two factors: (i) local Mn-rich crystallization environment, possibly due to the absence of spessartine-rich garnet, and (ii) a relatively high fractionation level of the pegmatite. In contrast, the TVB schorl composition indicates a simple evolution stage and low fractionation level of its parent pegmatite.

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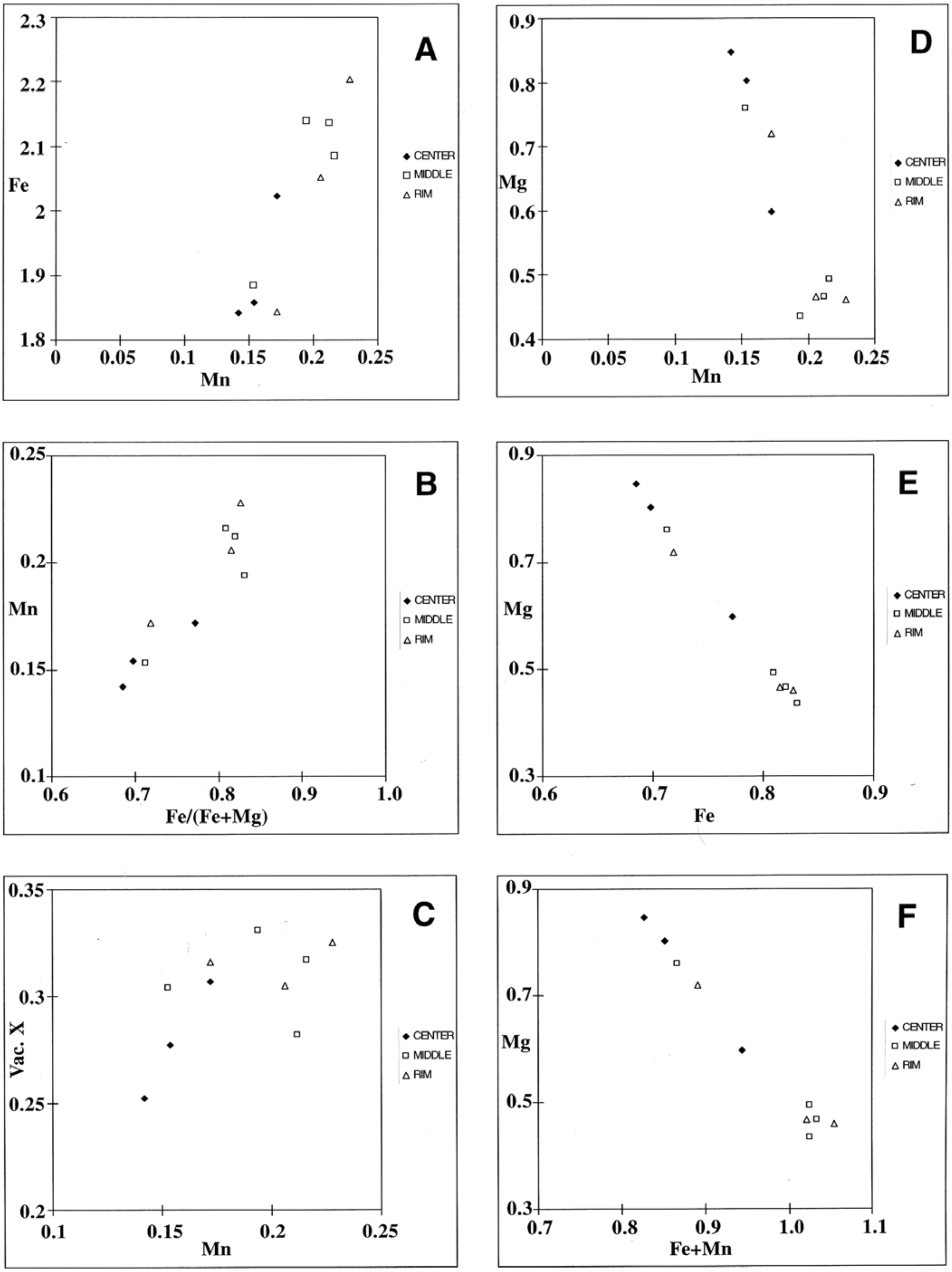

Fig. 2 Compositional diagrams of the Magurka, Adolf adit schorl (atoms per formula unit).

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Složení skorylu z primitivních granitických pegmatitů Západních Karpat: dva příklady z Nízkých Tater, střední Slovensko

Studium turmalínu na elektronové mikrosondě ze dvou primitivních granitických pegmatitů v Nízkých Tatrách, střední Slovensko, poskytlo tyto výsledky: (1) Studované vzorky turmalínu odpovídají skorylu, bohatému Al (hlavně vzorek TBV), s kolísajícím obsahem Mg, s poměry Fe/(Fe+Mg) = 0.54–0.60 a 0.69–0.83, nízkými koncentracemi Ti, Ca a K, a s vakancemi v pozici X (~20–30 atom. %). (2) Krystaly skorylu jsou slabě zonální se zvyšujícím se obsahem Fe a poměrem Fe/(Fe+Mg) od středu k okraji. (3) Neobvykle vysoké obsahy Mn zjištěné ve skorylu TMA jsou netypické pro primitivní granit-pegmatity. Vysoká koncentrace Mn, zvyšování poměru Fe/(Mn+Fe) a obsahu Fe, a pokles Mg směrem k okraji krystalu pravděpodobně odrážejí vliv dvou faktorů: (i) prostředí lokálně obohacené Mn, zřejmě způsobené absencí spesartinového granátu, a (ii) relativně vysokou úroveň frakcionace v pegmatitu. Na druhé straně, složení skorylu TVB naznačuje jednoduchý vývoj a nízkou úroveň frakcionace mateřského pegmatitu.